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## **Effect of Mechanical dry Coating on the Flowability and the Wettability of Silica Gel Powder**

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### **Abstract**

Dry particle coating has been used to create new-generation materials by combining different powders exhibiting different physical and/or chemical properties. Particles with relatively large particle size (host particles, 1-500  $\mu\text{m}$ ) can be mechanically coated with fine particles (guest particles, 0,1-50  $\mu\text{m}$ ) in order to create new functionality or to improve their initial characteristics.

In this study the dry coating experiments were performed with two different mixing processes using Hybridizer (Nara) and Cyclomix (Hosokawa). Silica gel particles ( $d_{50} = 55 \mu\text{m}$ ) were coated with fine particles of magnesium stearate ( $d_{50} = 4,6 \mu\text{m}$ ) at two different mass ratios of magnesium stearate, 5% and 15%. Several methods of characterization were used to study the effect of dry coating on the flowability and the wettability of silica gel powder.

The uncoated and coated silica gel particles were observed by environmental scanning electron microscopy (ESEM). The images showed that a more uniform coating was obtained in the case of Hybridizer. The flowability of the different samples obtained with Hybridizer and Cyclomix was characterized by measurements of the tapped and aerated densities. It has been shown that the flowability of silica gel treated in Hybridizer was not significantly affected whereas the flowability was reduced after treatment in the Cyclomix. The wettability of silica gel powder was determined by measurements of the contact angle between the water drop and the powder bed prepared for each sample. The results obtained showed that the coating of silica gel powder by hydrophobic magnesium stearate in both the Hybridizer and Cyclomix, improves its moisture resistance.

### **Keywords:**

*Dry coating ; Hybridizer ; Cyclomix ; characterization ; flowability ; wettability.*

### **I. Introduction**

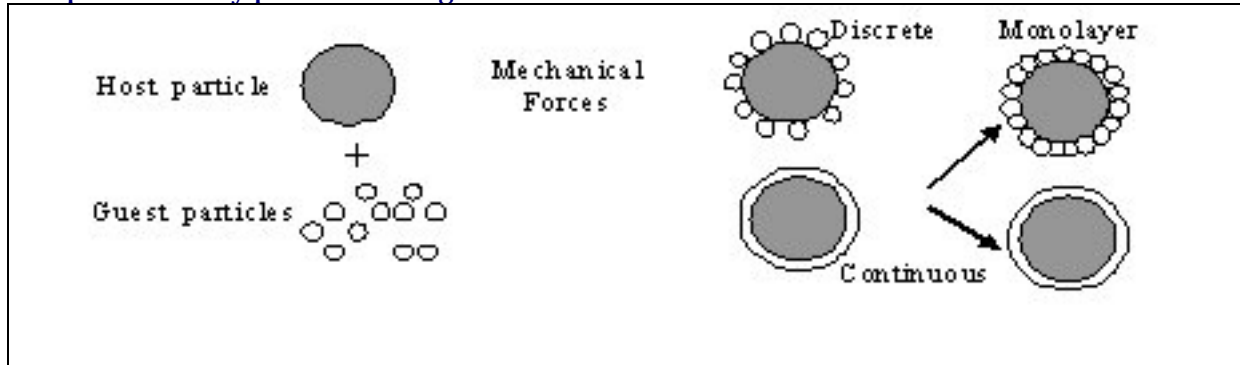
Dry particle coating to change the surface properties and/or functionality of powders appears as a very important for many industries. Typical applications include modification of flowability, wettability (hydrophobic/hydrophilic properties), solubility, dispersibility, flavour, shape, electrostatic, optical, electric, magnetic, particle properties.

In dry particle coating processes, materials with relatively large particle size (host particles; 1-500  $\mu\text{m}$ ) are mechanically coated with fine particles (guest particles; 0,1-50  $\mu\text{m}$ ) in order to create new functionality or to improve their initial characteristics (Yoshihara and Pieper, 1999). Since the size of the guest particles are so small, van der Waals interactions are strong enough to keep them firmly attached to the host particles. Thus, either a discrete or

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continuous coating of guest particles can be achieved depending on a variety of operation conditions including processing time, rotation speed, weight fraction of guest to host particles and particle properties (Pfeffer *et al.*, 2001). Figure 1 below is a simple schematic illustrating the process of dry particle coating.



*Figure 1. Dry particle coating principle*

The subject of dry coating is very closely related to the subject of dry mixing powders. In ordered mixing (a term coined by Hersey, 1975), the surface of larger particles is loosely coated with smaller particles. In dry particle coating, the same thing happens; however, the surface covering is more permanent because of a stronger physical (or chemical) bonding (Pfeffer *et al.*, 2001). Initial work on ordered mixing was done by Hersey and co-workers (Hersey, 1975; Hersey, 1977; Yeung and Hersey, 1979; Yip and Hersey, 1977a; Yip and Hersey, 1977b; Yip and Hersey, 1977c). The concept of ordered mixing was also taken one step further (to dry coating) by using dry impact blending, as described in a series of papers by Japanese group (Honda *et al.*, 1987; Honda *et al.*, 1988; Honda *et al.*, 1989; Honda *et al.*, 1991). Several different dry coating machines have been developed allowing the creation of new types of materials. (Pfeffer, 2001) gives descriptions of these different dry coating systems. (Mujumbar *et al.*, 2004) have studied dry coating to enhance the moisture resistance of ground magnesium powder by coating its surface with caruba wax. Coating was done using Magnetically Assisted Impaction Coating (MAIC) and two high-speed impaction devices, the Hybridizer (HB) and Mechanofusion. (Yang *et al.*, 2005) showed that it is possible to improve the flowability of cornstarch by coating with nanosized silica, using dry coating devices such as MAIC and HB.

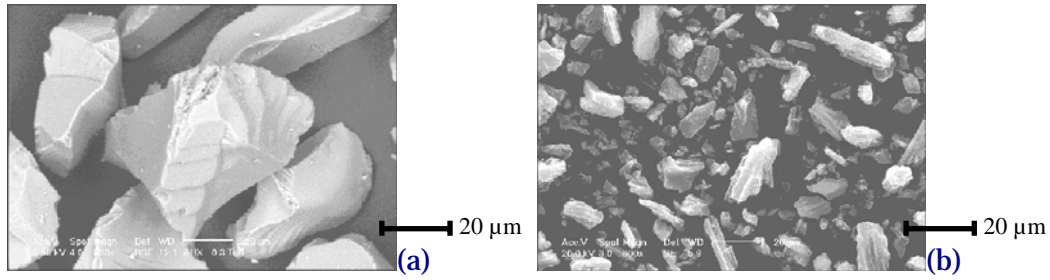
In this paper we describe an experimental investigation of an application of dry coating technique to study the effect of mechanical dry coating on the flowability and the wettability of silica gel particles coated with two different weight ratio of magnesium stearate by using a high energy impact blending coater “Nara Hybridizer” and a high shear mixer “Cyclomix”.

## **II. Experimental**

### **II.1 Powders**

Silica gel powder supplied by Merck and usually used for the filling of columns of chromatography has been chosen as host particles for dry coating. The silica gel is sensitive to the variations of humidity because its porous structure. As shown in Figure 2(a), the environmental scanning electron microscope (ESEM) image indicates that the silica gel particles are irregularly shaped and the surface is rough.

Hydrophobic magnesium stearate (MgSt) supplied by Chimiray is used as guest particles. The MgSt is a fine, white, greasy and cohesive powder widely used in pharmaceutical formulation as a lubricant. A ESEM image (Figure 2(b)) shows a very large distribution of size and shape including the needle and plate configurations. The properties of host and guest particles used in the experiments are summarized in Table 1.



**Figure 2. ESEM images of host and guest particles. (a) Silica gel; (b) MgSt**

**Table 1. Properties of host and guest particles**

| Particles  | Size ( $d_{50}$ ) ( $\mu\text{m}$ )<br>(Mastersizer 2000) | True density ( $\rho$ )<br>(g/cm <sup>3</sup> ) (Helium Pycnometer) | Specific surface area<br>( $S_{\text{BET}}$ ) (m <sup>2</sup> /g)<br>(Micromeritics ASAP 2010) |
|------------|---|---|--|
| Silica gel | 55 ( $D_{\text{host}}$ )                                  | 2,07 ( $\rho_{\text{host}}$ )                                       | 510  |
| MgSt       | 4,6 ( $d_{\text{guest}}$ )                                | 1,04 ( $\rho_{\text{guest}}$ )                                      | 6  |

## II.2 Coating processes

The evolution of the percentage by mass of guest particles used in coating experiment is calculated based on the assumption of 100% surface coverage of the host particles with a monolayer of guest particles. We assume that all guest particles are of same size, both host and guest particles are spherical, and that the shapes of host and guest particles do not change during the coating process. Based on these assumptions, the mass percentage ( $W$ ) of guest particles for 100% coverage can be written as (Yang *et al.*, 2005):

$$W(\%) = \frac{(Nd_{\text{guest}}^3 \rho_{\text{guest}})}{(D_{\text{host}}^3 \rho_{\text{host}}) + (Nd_{\text{guest}}^3 \rho_{\text{guest}})} \times 100 \quad (1)$$

For  $D_{\text{host}} \gg d_{\text{guest}}$  (here,  $D_{\text{host}} / d_{\text{guest}} \approx 10$ ), the number  $N$  of guest particles per host particle is given by the expression:

$$N = \frac{4(D_{\text{host}} + d_{\text{guest}})^2}{d_{\text{guest}}^2} \quad (2)$$

From Equation (1), the percentage of guest particles needed to silica gel particles is 15%. In addition, coating experiments have been carried out with 5% of magnesium stearate.

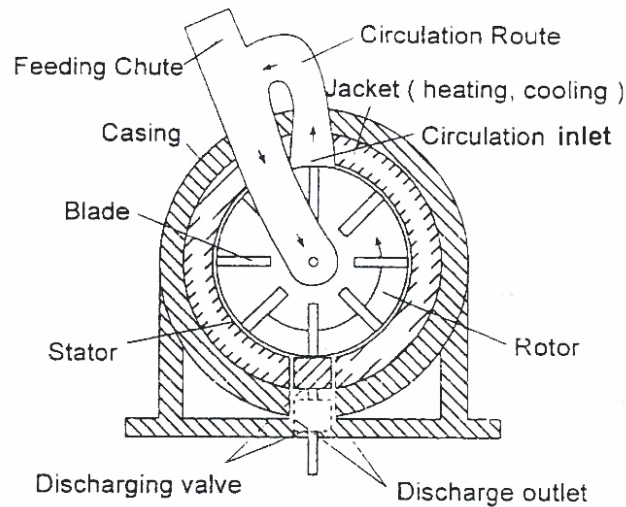
Two different dry coating devices were studied to determine the coating performance as described below:

### II.2.1 Hybridization system (HB, type NHS-0; Nara Machinery Co.)

It is a high-speed dry impact blending coater, which comprises an OM. Dizer, the hybridizer, the product collector and the control panel (Yoshihara, 1999; Pieper, 1996). Figure 3 is a schematic diagram of the hybridizer. It consists of a very high-speed rotating rotor with six blades, a stator and a powder re-circulation circuit (Yang *et al.*, 2005). The coating chamber is surrounded with a jacket in which coolant is circulated (Kangwabtrakool and Shinohara, 2001).

The processing can be summarized as follows: the powder mixture (host and guest particles) is subjected to high impactation and dispersion due to collisions with blades and the walls of the device and continuously re-circulates in the machine through the cycle tube. Particle coating is achieved due to the embedding or filming of the guest particles onto the host particles by high impactation forces and friction heat (Yang *et al.*, 2005). Since the rotor of the hybridizer can rotate anywhere from 5000 to 16000 rpm, very short processing time is

required to achieve coating (Pfeffer *et al.*, 2001). The operating conditions used in experiments are 4800 rpm for 5 min.



*Figure 3. Design of the hybridizer apparatus (Kangwabtrakool and Shinohara, 2001)*

### *II.2.2 Cyclomix (Hosokawa Micron B.V.)*

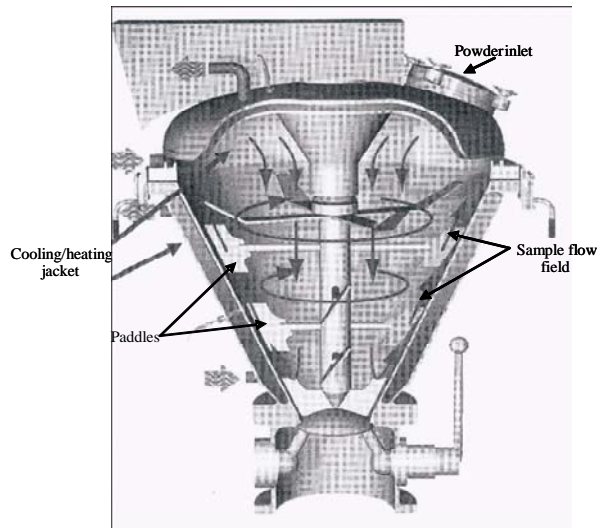
This model of vertical high shear mixer granulator provided by Hosokawa Micron presents and has a nominal volumic capacity of 1 L. The mixer has four pairs of fat-bladed impellers from the bottom to the top. A centrally located high-speed rotating shaft is driven from the mixer cover, thus eliminating seals and bearings from the product zone. The shaft is fitted with paddle-shaped mixing elements, which rotate close to the inner vessel wall (Ng *et al.*, 2007). Figure 4 is a schematic diagram of the Cyclomix mixer. The working principle of the Cyclomix differs markedly from the existing mixing techniques owing to the specific interaction between mixing element and vessel wall. The powder (host and guest particles) is loaded into the conical mixing vessel from the top; the degree of filling can range between 30 and 100%. Together, the high-speed rotation (up to 2500 rpm) of the paddles and the conical shape of the vessel force the product from the bottom to the upper zone of the vessel. Upon reaching the top, the product flows downwards into the centre of the vessel. This flow pattern results in fast macromixing. During the upward motion, the particles are accelerated by the paddles and intensively mixed by friction with vessel walls. The operating conditions used in experiments are 1500 rpm for 5 min.

## **III. Characterization**

The uncoated and coated silica gel particles were examined by means of an environmental scanning electron microscopy (ESEM) to study the surface morphology and particle shapes before and after coating. The tapped density tester (Volumeter) has been used to examine the flowability of the uncoated and coated samples. The tapped density is obtained by mechanically tapping a powder sample contained in a measuring cylinder. After observing the initial volume, the cylinder is mechanically tapped, and volume readings are taken until little volume change is observed. The flow properties of the uncoated and coated powders were evaluated by the Carr's flowability index (FI) and Hausner's ratio (HR).

The wettability has been studied by the sessile drop method (Lazghab *et al.*, 2005; Goalard, 2005). A small water drop of 10  $\mu$ l is deposited on the surface of a powder bed prepared for each sample. The shape of the drop profile is observed and used to determine the contact angle.



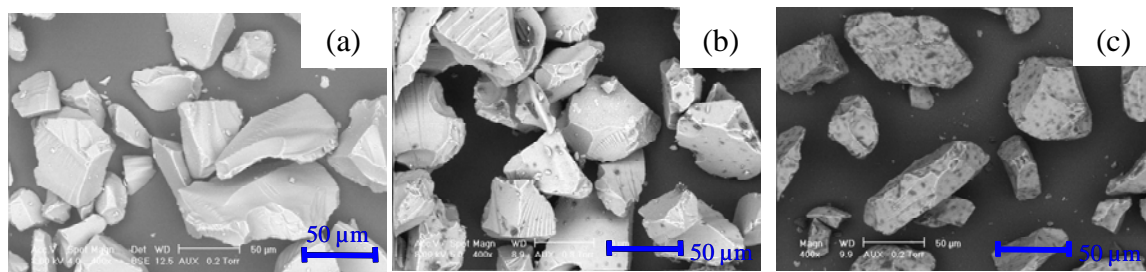


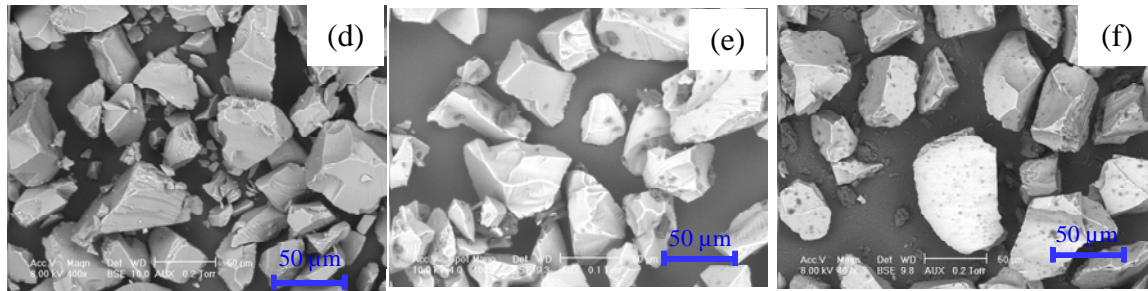
*Figure 4. The Cyclomix high shear mixer (Kwan et al, 2005)*

## IV. Results and discussion

### IV.1 Surface morphology

The surface morphology of particles processed alone and coated with 5% and 15% (w/w) of MgSt in the Hybridizer and the Cyclomix processes is shown in Figure 5. The ESEM images were carried out just after treatment in both the Hybridizer and Cyclomix because of the coated silica gel powder ageing observed in fewer 10 days. The particle of silica gel in its original form (Figure 2(a)) is irregularly shaped. The shape of particles was not changed by the hybridizer treatment (Figure 5(a)) but after processing in Cyclomix, the silica gel particles were crushed probably because of high shearing in the mixer (Figure 5(d)). The MgSt guest particles were discretely distributed on the surface after treatment in both the Hybridizer and the Cyclomix. The MgSt has been softened and smeared over the silica gel particles (Figures 5(c), 5(f)). Greater MgSt coverage is observed on the surface of silica gel as the MgSt percentage is increased to 15%. Differences in the morphology of the MgSt coating in the two different devices are observed. More MgSt can be seen on the particles when they are processed in the Hybridizer due to the higher impact forces and higher local temperature in this device (Figure 5(c)). Much more free fine particles of MgSt were observed in the product from Cyclomix process (Figure 5(f)).





*Figure 5. SEM images of Silica gel particles treated in Hybridizer: alone (a); coated with 5% (w/w) of MgSt (b); coated with 15% (w/w) of MgSt (c) and in Cyclomix: alone (d); coated with 5% (w/w) of MgSt (e); coated with 15% (w/w) of MgSt (f)*

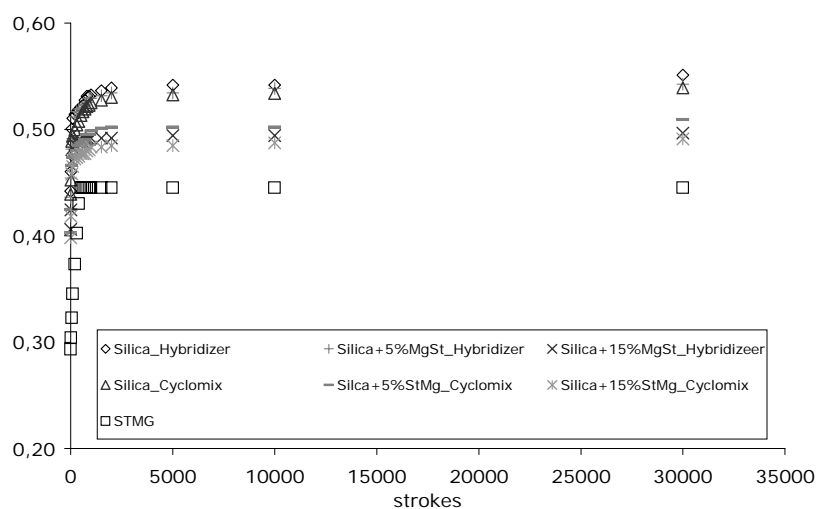
#### IV.2 Flowability tests

The as-received silica gel powder presents good flow properties. The flowability of silica gel processed alone in Hybridizer is not affected and remains good enough whereas the treatment in the Cyclomix reduces the flow properties because of the grinding of particles. The values of the Carr's flowability index (FI) and Hausner's ratio (HR) of uncoated and coated silica gel particles and MgSt are summarized in Table 2.

Figure 6 shows the variation of the tapped density measured for the silica gel processed alone or coated with MgSt in both the Hybridizer and Cyclomix. One can observe that the flowability of silica gel becomes mediocre after coating with 5% of MgSt in Cyclomix and decreases strongly with 15% of MgSt. For the particles treated in Hybridizer, the flow properties remain good for the coated particles with 5% of MgSt and decreases slightly for the weight percentage of 15% of MgSt.

*Table 2. The values of Carr's flowability index (FI) and Hausner's ratio (HR)*

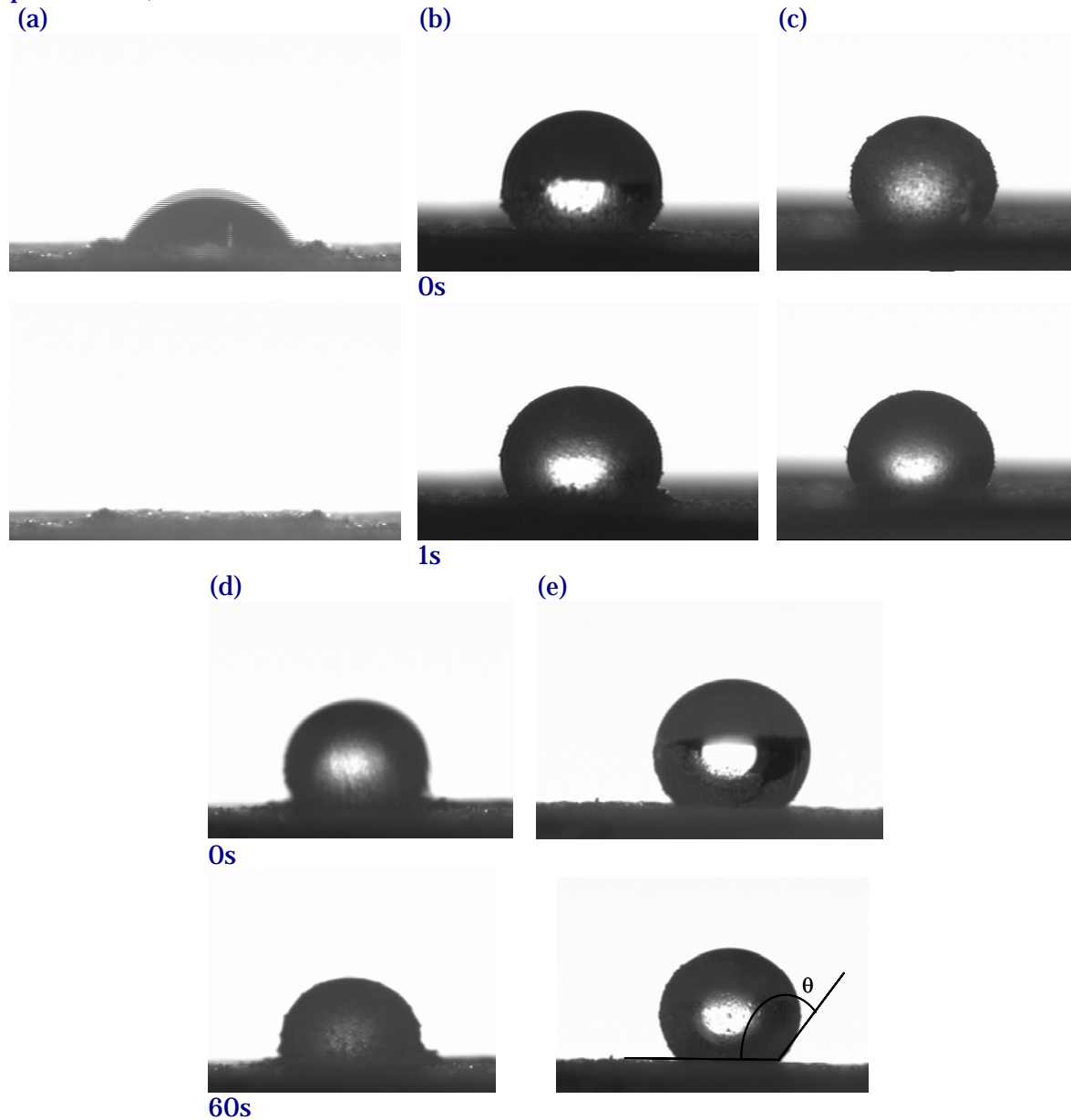
| Samples               | FI    | HR   | Flowability |
|-----------------------|-------|------|-------------|
| Silica gel_Hybridizer | 15,24 | 1,18 | good        |
| Silica gel_Cyclomix   | 18,45 | 1,23 | mediocre    |
| 5%MgSt_Hybridizer     | 17,60 | 1,22 | good        |
| 5%MgSt_Cyclomix       | 20,96 | 1,27 | mediocre    |
| 15%MgSt_Hybridizer    | 18,22 | 1,22 | mediocre    |
| 15%MgSt_Cyclomix      | 21,21 | 1,27 | bad         |
| MgSt                  | 34,10 | 1,52 | bad         |



*Figure 8. Flowability of uncoated and coated host particles*

### IV.3 Wettability tests

Since the MgSt is hydrophobic, the coating should make the hydrophilic silica gel surface become hydrophobic. To evaluate the impact of coating on the reduction of the high affinity between silica gel and water, wettability tests were carried out for the coated silica gel particles in both the Hybridizer and Cyclomix and the uncoated particles. The results of the measured contact angle (at 0s) and the work of adhesion (Wadh) describing the interaction between two different phases (the required work to extract the water drop from the surface of powder bed) are summarized in Table 3.



*Figure 9. Wettability of Silica gel powder before coating: (a), after processing in the Hybridizer: (b) 5%MgSt; (c) 15%MgSt and in the Cyclomix: (d) 5%MgSt; (e) 15%MgSt*

Figure 9 shows photos taken after two different times of contact between the water drop and the solid surface. The tests have been carried out on powder beds of silica gel and coated particles with 5 and 15% of MgSt in both the Hybridizer and Cyclomix. It can be seen that the



water drop disappears instantaneously (after 1s) on the surface of silica gel powder (Figure 9 (a)) because of the high affinity between silica gel and water.

*Table 3. Results of the measured contact angles and the work of adhesion*

| Samples            | Average contact angle<br>( $\theta^\circ$ ) | Wadh (mN<br>$m^{-1}$ ) |
|--------------------|---|------------------------|
| Silica gel         | 15,2  | 1,2                    |
| 5%MgSt_Hybridizer  | 112   | 46                     |
| 5%MgSt_Cyclomix    | 98  | 62                     |
| 15%MgSt_Hybridizer | 116   | 41                     |
| 15%MgSt_Cyclomix   | 127   | 29                     |
| MgSt               | 125   | 32                     |

After coating by 5 and 15% MgSt in the Hybridizer (Figures 9 (b) and (c)) and in the Cyclomix (Figures 9 (d) and (e)), the water drop is not absorbed and remains on the surface of silica gel powder. The measured contact angle increases when the MgSt percentage increases whereas the work of adhesion decreases (Table 3). These results clearly prove on the one hand that the surface of the silica gel particles is coated by the MgSt particles and on the other hand that the affinity of the silica gel powder with water is enhanced by dry coating in both the Hybridizer and Cyclomix.

## V. Conclusions

This study indicates that it is possible to change the properties of silica gel particles by coating with hydrophobic magnesium stearate using both the hybridizer and Cyclomix as dry coating devices. The ESEM images of the uncoated and coated particles in the two devices show that MgSt was softened and smeared over host particles but the silica gel treated alone in the Cyclomix was crushed whereas no difference was observed after processing in the Hybridizer. The flowability of the silica gel powder was not strongly affected by coating in the Hybridizer and remains good. After treatment in the Cyclomix, the flow properties of silica gel are significantly decreased probably because of the grinding of particles. It is also found that the coating by hydrophobic MgSt in both the Hybridizer and Cyclomix can also reduce the high affinity between silica gel and water.

In summary, it has been demonstrated that a dry particle coating technique can be used to modify the properties of silica gel powder by coating with small quantities of hydrophobic magnesium stearate.

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